

# Microwave-Enhanced hopping-conductivity; a non-Ohmic Effect

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## Abstract

Hopping conductivity is enhanced when exposed to microwave fields (Phys. Rev. Lett., **102**, 206601, 2009). Data taken on a variety of Anderson-localized systems are presented to illustrate the generality of the phenomenon. Specific features of these results lead us to conjecture that the effect is due to a field-enhanced hopping, which is the high frequency version of the non-Ohmic effect, well known in the dc transport regime. Experimental evidence in support of this scenario is presented and discussed. It is pointed out that existing models for non-Ohmic behavior in the hopping regime may, at best, offer a qualitative explanation to experiments. In particular, they cannot account for the extremely low values of the threshold fields that mark the onset of non-Ohmic behavior.

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## I. INTRODUCTION

Non-Ohmic effects are commonly encountered in hopping conductivity. Actually, it is difficult to maintain linear-response conditions in these systems. The degree of non-Ohmicity could be tamed by reducing the potential drop across the sample but the need to ensure good signal-to-noise makes this harder as the temperature gets lower. Deviations from linear response may be expected when the applied field  $F_c$  obeys:

$$F_c \gtrsim \frac{k_B T}{eL} \quad (1)$$

Here,  $k_B$  is Boltzmann constant;  $e$  is the electron charge, and  $T$  the temperature.  $L$  is the spatial scale over which the electron gains energy from the field  $F$  before it is dissipated into the bath, usually by phonon emission, and therefore  $L$  is a function of temperature, typically of the form  $L \propto T^{-p}$  ( $p$  is a number between 1 to 4 depending on the material, temperature range, and dimensionality). Modifications to the conductance by a sufficiently large dc field were studied by several, somewhat different theoretical approaches<sup>1-4</sup>. These models however predicted a similar result; the field induces an excess conductance  $\Delta G$  that, at intermediate field-strength, is exponential with  $(eFL/k_B T)^\gamma$  with  $\gamma=1$  or 2. Intermediate fields are fields in the range:  $F_c < F < F_h$ . The high-field limit  $F_h$  (where the conductance becomes independent of temperature<sup>4,5</sup>), is defined by  $F_h \gtrsim \frac{k_B T}{e\xi}$ , where  $\xi$  is the localization length.

Much less attention has been given to the effect of a long-wavelength electromagnetic radiation on hopping conductivity. Of particular interest is where the wavelength is larger than the sample-size<sup>6</sup> which can be readily implemented by using microwaves (MW) radiation. Ben-Chorin et al observed that exposing the sample to a MW source enhanced its hopping conductance and, perhaps naturally, associated the effect with heating<sup>7</sup>. However, systematic studies, exploiting some unique properties of electron-glasses<sup>8</sup>, suggested that this is a non-equilibrium effect. A persistent feature of the results,

which made it difficult to reconcile the effect with heating, is a sub-linear dependence of  $\Delta G$  on the microwaves power  $P$ .

In this paper we report on results obtained by similar measurements on films of five different hopping systems; granular-aluminum,  $\text{In}_2\text{O}_{3-x}$  (crystalline indium-oxide),  $\text{In}_x\text{O}$  (amorphous indium-oxide), beryllium, and GaAs. In all cases  $\Delta G$  at  $T \approx 4\text{K}$  turns out to be sub-linear with the MW power in agreement with the results of previous studies. It is further shown that the functional dependence of  $\Delta G(P)$  is consistent with the current-voltage characteristics measured independently (at low frequency) on the same samples. The MW-enhanced conductance is then conjectured to be a non-Ohmic effect, of a similar nature as the field assisted-hopping phenomenon. This is a generic mechanism and should be obeyed by any system once driven away from its linear response transport regime.

As a further test of our conjecture, the temperature dependence of the MW-induced  $\Delta G$  is shown to be in qualitative agreement with models for field assisted hopping. On the other hand, a quantitative analysis of these non-Ohmic effects exposes a discrepancy between these theories and experiments. This discrepancy, well documented in experiments on hopping systems, manifests itself as a much higher sensitivity to fields than suggested by equation 1. It is also pointed out that the exponential dependence on the field, anticipated by theory, might hold only over a very small range of fields.

## II. EXPERIMENTAL

Several materials were used to prepare samples for measurements in this study. These were thin films of  $\text{In}_2\text{O}_{3-x}$  and  $\text{In}_x\text{O}$ , and granular-aluminum. Samples from these materials were prepared by e-gun evaporation of  $\text{In}_2\text{O}_3$  or aluminum pellets unto room temperature glass substrates in partial oxygen pressures of  $(4-6) \cdot 10^{-5}$  mbar and rates of  $(0.3-1)$  Å/s. The  $\text{In}_2\text{O}_{3-x}$  samples were obtained from the as-deposited amorphous films by crystallization at 525 K. We also used for comparison samples of GaAs and beryllium that were highly

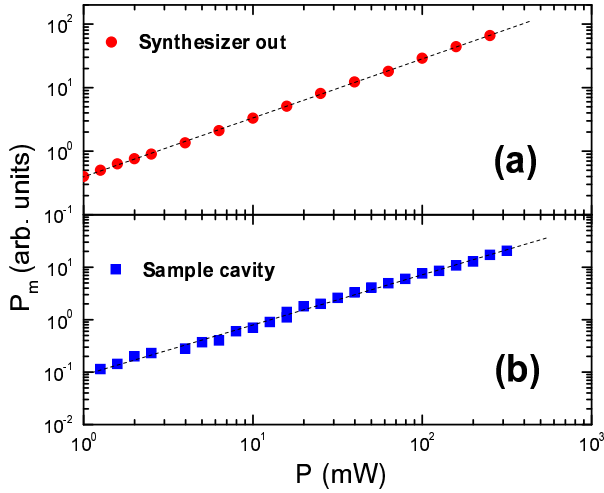


FIG. 1: The power  $P_m$  recorded by the power-meter as function of the output power setting on the synthesizer  $P$ . The latter is used throughout the paper to characterize MW power. Measurements shown are at a constant frequency of 2.45 GHz; (a) measured about 2 cm from the synthesizer output socket. (b) measured 1 cm from the short antenna at the sample-end of the transmission line.

resistive and exhibited hopping conductance at liquid helium temperatures.

Conductivity of most of the samples was measured using a two terminal ac technique employing a 1211-ITHACO current preamplifier and a PAR-124A lock-in amplifier. The GaAs specimen however, was configured for a Van-der-Pauw, 4-terminal ac measurement. In either case, the ac voltage bias was kept small enough to minimize deviations from Ohmic conditions (as will be shown below, some deviation may be observed even at the smallest fields used).

The high-power synthesizer HP8360B, was used with power up to 25 dBm ( $\approx 316$  mW) for the MW excitation. The frequency range used in this work was limited to 2-6 GHz. The output of the synthesizer was fed to the sample chamber via a coaxial cable ending with a short antenna. An important issue in the study is the functional form of the sample response to the power of the MW. It was therefore desired to ascertain the linearity of the synthesizer setting versus its output as well as the integrity of the transmission line itself. This was done by measuring the generated power near the instrument output and near the sample stage. The results shown in figure 1 seem to rule out the possibility that the sub-linearity is an instrumental artifact.

Some auxiliary measurements described below employed the Tabor WS8101, a 100 MHz generator as the radio-frequency (RF) source. The output of this generator was inserted into the sample cavity through the same transmission line as the MW source. The linear relation between the setting of the generators and the induced RF voltages across the sample was ascertained by mon-

itoring the pick-up waveforms on an oscilloscope. The data shown in this paper are plotted with respect to the output settings of the respective RF or MW source.

In both the MW and RF experiments an initial frequency scan was made to locate a range where the response is conveniently large. Naturally that was usually one of the resonances of the chaotic cavity in which the sample was mounted. As will be shown below, the positions of these resonances was stable over many hours, and reproducible results could be obtained with a barely noticeable drift in frequency ( $<0.1\%$ /day).

Unless otherwise mentioned, measurements reported here were performed at  $T \approx 4.1$  K with the sample immersed in liquid helium. Complementary details of sample preparation, characterization, and measurements techniques are given elsewhere<sup>9</sup>.

### III. RESULTS AND DISCUSSION

The most characteristic feature of the MW-enhancement phenomenon is the functional form of the excess conductance  $\Delta G$  versus the radiation power  $P$ . In more than 150 samples studied in our laboratory,  $\Delta G$  increased with  $P$  less than linearly. Results of  $\Delta G(P)$  scans, illustrating the sub-linear form of the response, are shown in figure 2. The sub-linearity in these results are typical; cases where the sub-linearity was less prominent than those in figure 2 were encountered usually only in samples where the maximum effect was smaller than 2 %.

While linearity at power levels smaller than measured here cannot be ruled out, it is noted that there are instances where  $\Delta G(P)$  is sub-linear at a power such that  $\Delta G/G \leq 10^{-2}$ . This was the first indication that the origin of the effect is not consistent with 'heating'; from the point of view of power-balance, a spatially uniform increase of the electron temperature will perforce give  $\Delta G \propto P$  when both  $\Delta T$  and  $\Delta G$  are small as is evidently the case for  $\Delta G/G \leq 10^{-2}$ . An even stronger evidence against 'heating' was given in<sup>8</sup> based on the lack of change in the 'memory-dip' shape upon exposure to MW radiation. The memory-dip is an identifying feature of intrinsic electron-glasses. It is a cusp-like modulation observable in conductance versus gate voltage sweeps<sup>10</sup>. It has a shape that is highly sensitive to the electron temperature and thus may be used a thermometer (see<sup>10</sup> for details). The underlying mechanism for the MW-enhanced conductivity was not identified in<sup>10</sup>.

As more data became available, a statistical correlation emerged between the sensitivity of  $G$  to the voltage employed in the conductance measurement, and the magnitude of  $\Delta G$  produced by a given power of MW; samples that showed Ohmic behavior up to relatively high voltages, exhibited small MW-induced  $\Delta G$ , and *vice-versa*.

The connection between the two effects became clear once their functional form was compared. To affect this comparison the  $\Delta G(P)$  is converted to  $\Delta G(b \cdot P^{1/2})$  where

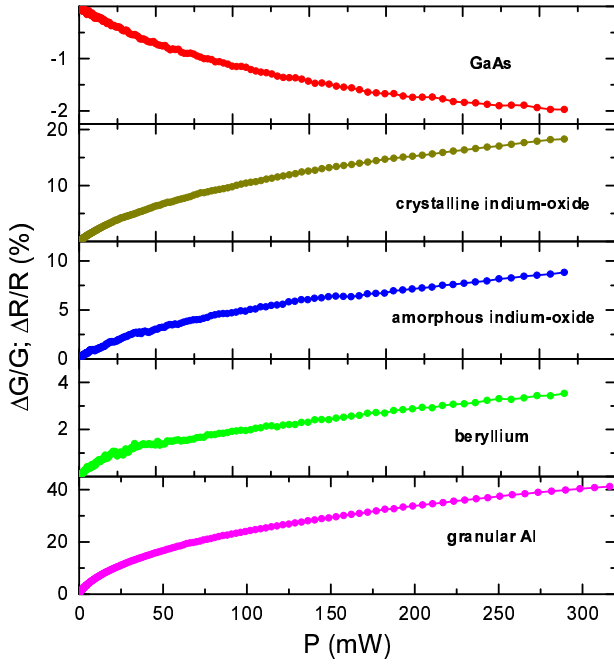


FIG. 2: The fractional change of conductance (resistance in the case of the GaAs sample, which was measured by a 4-terminal technique), as function of MW power. All samples shown were measured at  $T=4.1\text{K}$ .

$b$  is a constant chosen to make an optimal fit to  $\Delta G(F)$  obtained independently from the current-voltage characteristics of the sample. The similarity between the two functions, was observed in all the tested samples, examples are shown in figure 3 and 4 below.

The following heuristic picture for the MW enhancement mechanism then suggests itself: The MW radiation induces field across the sample<sup>11</sup> (picked-up by the wires connected to the sample for the two-terminal conductance measurement). Once the potential difference associated with this field is greater than the voltage employed for measuring  $G$ , the conductance of the sample is modified in a similar vein as in measuring  $G(F)$  directly (by dc or low-frequency ac technique). In other words, the conductance monitored by a low-bias, low-frequency technique takes advantage of the improved current-path induced by the high-bias associated with the MW field. The reason for the sub-linearity of  $\Delta G(P)$  is then just the less-than-quadratic dependence of the excess conductance on an applied field over the range relevant for the measured  $\Delta G$ .

$\Delta G(F)$  of a given sample is similar but it does not perfectly match the respective  $\Delta G(b \cdot P^{1/2})$  (see, for example, figures 3 and 4). There were always some degree of imprecise registry between these functions in all the samples we tested. Such deviations ought perhaps to have been expected. The assumption, implicit to our matching procedure, that it is only the *amplitude* of the field that matters, is inaccurate; the Miller-Abrahams<sup>12</sup>

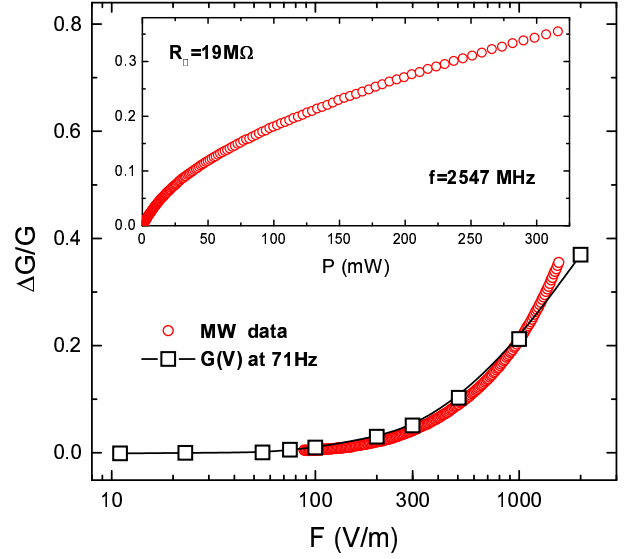


FIG. 3: The fractional conductance change of a 210 Å thick  $\text{In}_x\text{O}$  sample,  $1 \times 1 \text{ mm}^2$  as function of the applied field. This is compared with  $\Delta G(P^{1/2})/G$  by adjusting its x-axis (see text). The inset shows  $\Delta G(P)/G$  of this sample.

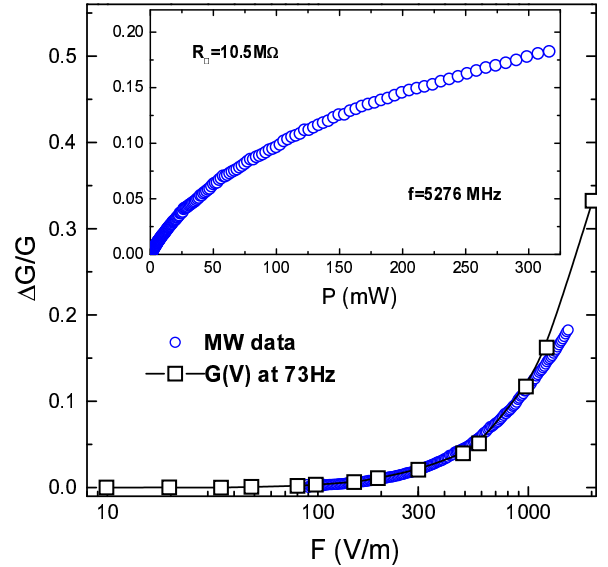


FIG. 4: Same as in figure 3 for a 50 Å thick  $\text{In}_2\text{O}_{3-x}$  sample.

resistors comprising the hopping system are not purely resistive, and therefore, the local potential drops across the sample may be different at different frequencies.

To elucidate this point, we measured  $\Delta G[F(\omega)]$  at different frequencies on several samples.  $F(\omega)$  was the modulating field applied at frequency  $\omega$  while the conductance was monitored at some low-frequency (typically, 20-75 Hz) as in the MW experiments. Here however, we employed frequencies in the  $10^6$ - $10^8$  Hz range so the voltages induced across the sample could be readily displayed

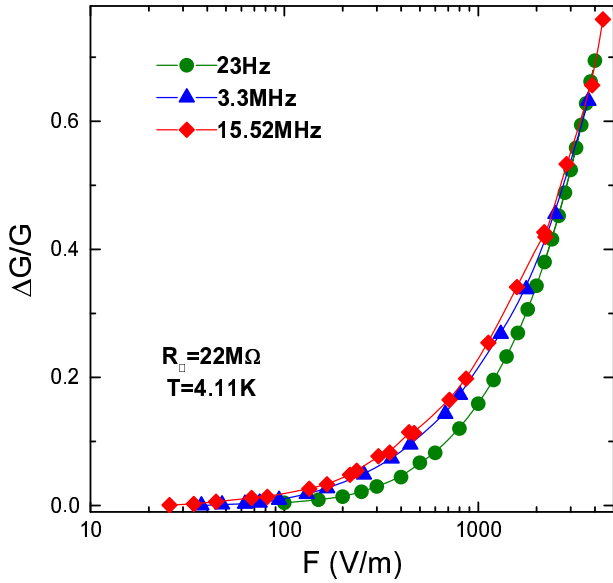


FIG. 5: The fractional change of conductance as function of field (measured at 23 Hz by a two-terminal lock-in technique). The two other curves were measured on the same sample, by exposing it to radiation at the indicated frequency. The field-axis of the high frequency curves was adjusted to match the 23 Hz curve at  $\Delta G/G=0.7$ . The sample is  $\text{In}_2\text{O}_{3-x}$  with thickness 43 Å.

and measured by an oscilloscope. Figure 5 shows results of such an experiment. The 3.3 MHz and 15.52 MHz curves in the figure were adjusted to coincide with the 23 Hz plot at  $\Delta G/G=0.7$  by re-scaling their voltage axis by a constant, just as in the MW case. This illustrates that the  $\Delta G(F)$  is also a function of the field frequency. Interestingly, above a certain frequency the constant used for re-scaling the  $\Delta G[F(\omega)]$  curves was somewhat smaller as  $\omega$  became higher. Apparently, to achieve the same  $\Delta G/G$ , larger amplitude is needed at higher  $\omega$ . This issue is currently under investigation.

The conjecture that the MW-enhanced conductance is a field-induced non-Ohmic effect is also supported by the temperature dependence of the excess conductance (at constant MW power). In these experiments,  $\Delta G(\omega)$  was measured over a range of frequencies at each temperature. This was done to cater for drifts in the cavity characteristics with the concomitant shift in conductance-response peaks (see figure 6). The actual drift observed in the figure is remarkably small; note that the series of plots were collected over 3 days because at each temperature the sample was allowed to equilibrate for several hours (the sample is an electron-glass with slow relaxation times<sup>13</sup>).

The inset to the figure shows  $\Delta G(T)$  due to the MW radiation normalized by  $G(T)$  at zero field. These experimental points fit reasonably well a power-law which may actually be in qualitative agreement with theory: According to Pollak and Riess<sup>4</sup>, and Shklovskii<sup>3</sup>, at fields

just above the "Ohmic regime" the conductance versus field is given by:

$$G(F, T) = G(T) \cdot \exp \left[ \frac{eFL(T)}{k_B T} \right] \quad (2)$$

and therefore:

$$\frac{\Delta G(F)}{G} = \exp \left[ \frac{eFL(T)}{k_B T} \right] - 1 \quad (3)$$

For small  $\Delta G/G$  this yields:

$$\frac{\Delta G}{G} \propto \frac{eFL(T)}{k_B T} \quad (4)$$

Pollak and Riess associate  $L(T)$  with the hopping length<sup>4</sup> while Shklovskii associates it with the percolation radius<sup>3</sup>. In either case one expects a power law dependence as indeed observed (see, inset to figure 6). To push the analysis a little further, note that the resistance versus temperature of this sample exhibits Mott's variable-range-hopping<sup>14</sup> of a two-dimensional system (figure 7). In this case the hopping length  $r$  scales with temperature as  $r(T) \propto T^{-1/3}$  and the percolation-radius  $L_C$  scales like  $L_C(T) \propto T^{-2/3}$  giving  $\Delta G/G \propto T^{-1.33}$  and  $\Delta G/G \propto T^{-1.67}$  respectively. The fit to the data in the inset to figure 6 yields  $\Delta G/G \propto T^{-1.75}$  which may suggest that  $L_C$  is the relevant length.

The consistency with theory, as to temperature dependence, encouraged us to test the agreement of the theory<sup>3</sup> with the experiment in a more detailed way. Using the relevant parameters from the  $R(T)$  data of the sample (figure 7), we estimate the percolation-radius,  $L^* \approx \xi \cdot (T_0/T)^{2/3} \approx 5000$  Å at 4K ( $\xi$  is the localization-length). This value of  $L_C$  is used in equation 2a to plot  $\Delta G(F)/G$  and the result is compared with the experimentally measured values in figure 8.

This comparison brings to light several problems that need be addressed. The theory *may* fit the experiment for very small  $\Delta G(F)/G$ , however, it systematically deviates from the experimental curve when  $\Delta G(F)/G \gtrsim 6\%$ . This discrepancy is not due to the uncertainty in the value of  $L_C$ : The experimental curve is simply *not* exponential over the range where the effect is more than few percents, (which makes the use of this function questionable). Nor is the range of the studied fields out of the 'intermediate' regime. The field where the problem appears is still *much* smaller than the high-field regime, where the conductance becomes temperature independent<sup>5,15</sup>. On the other hand, if we accept that the fit from  $F \approx 1$  V/m to  $F \approx 80$  V/m (see inset to figure 8) is in agreement with theory, we have a problem accounting for the condition for the onset of the exponential dependence: This is expected to be given by equation 1, and inserting  $F \approx 1$  V/m gives  $L \approx 400$  μm. It is difficult to assign a physical meaning to such a length-scale in a hopping system at  $\approx 4$ K.

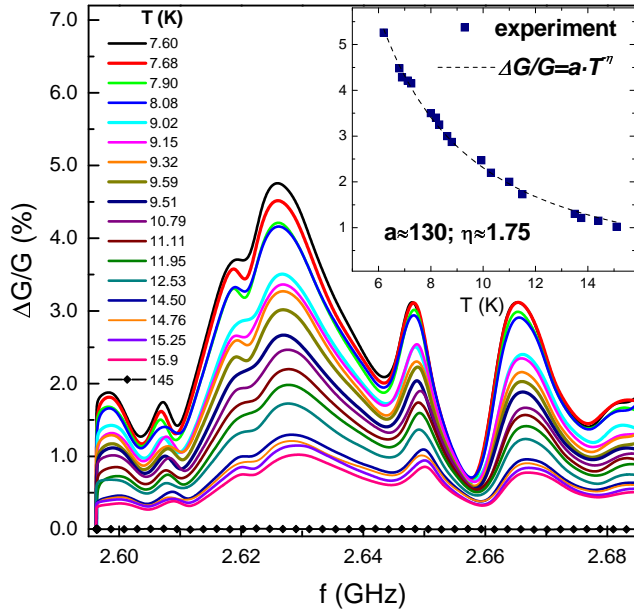


FIG. 6: Isotherm plots for the fractional change of conductance versus the MW frequency. Data are taken under a constant power of 25 dBm. The sample is 50 Å  $\text{In}_2\text{O}_{3-x}$  film. Note the shift of the  $\Delta G(f)$  peak positions with temperature. The inset shows the temperature dependence of  $\Delta G(f)/G$  averaged over the range:  $f=2628\pm 20$  MHz (to take care of the shift with temperature of the resonance peaks). The dashed line is a power-law fit to the data (c.f., equation 3).

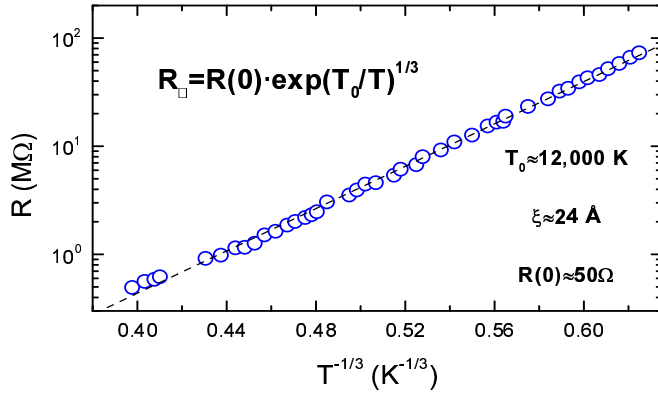


FIG. 7: Resistance as function of temperature for the same  $\text{In}_2\text{O}_{3-x}$  sample shown in figure 6.

This problem is commonly encountered in hopping conductivity. Usually, the puzzle is presented in terms of length-scale (being the one parameter in equation 1 that is not "measured"). Values of this length that are larger by 1-3 orders of magnitude than reasonably expected are reported, or could be inferred, from data in the literature<sup>16–18</sup>. These include results for Ge samples doped by nuclear-transmutation<sup>17</sup>, presumed to be relatively free of technical inhomogeneities. Remarkably, the

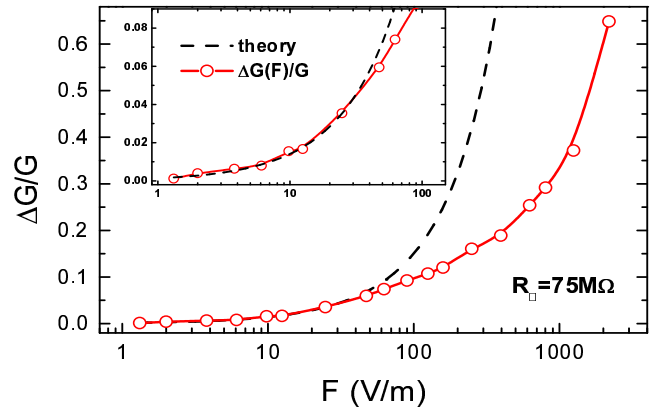


FIG. 8: The fractional change of the conductance as function of applied field for the  $\text{In}_2\text{O}_{3-x}$  sample shown in figure 6. The inset is a zoomed-in view of the small field region illustrating that non-Ohmicity is evident at very small fields. The "self-similar" impression one gets by comparing the zoomed-in view with the data in the main figure is a graphical testimony to that  $\Delta G(F)$  is not an exponential function.

field where non-Ohmicity was already evident in these experiments was smaller by at least<sup>17</sup> a factor of 25 than the theoretical value. These authors<sup>17</sup> find agreement with the Shklovskii model<sup>3</sup> in terms of the temperature dependence of  $L(T)$ , however, their data fit equation 2 only over a very small range ( $\leq 6\%$ ).

It has been remarked by several authors<sup>15,16</sup> that the origin of the discrepancy between theory and experiment as to the onset-field for non-Ohmicity may be a result of non-uniform field distribution. This might make the field effectively larger than the field one uses in equation 1 based on the measured potential difference across the sample divided by its length. Spatial inhomogeneity is inherent to Anderson insulators, and at least partially, this is actually embodied in the theory in that the system is self-averaging only on scales larger than  $L_C$ , which may be quite large at low temperatures. The observation that there has to be a larger length scale to account for the experimental results on different systems suggests that there is an inherent reason not taken into account in the standard hopping models (in addition to imperfections due to technological reasons). There seems to be evidence, based on studies of the onset of non-Ohmic behavior as function of sample-size<sup>19</sup>, that long-range potential-fluctuations may play a role in this phenomenon.

Note, incidentally, that the sub-linearity of  $\Delta G(P)$  at small fields follow naturally from theoretical models that predict  $G(F) \propto \exp[\frac{eFL}{k_B T}]$ <sup>1,3,4</sup>. The Apsley and Hughes model<sup>2</sup> that expects  $G(F) \propto \exp[(\frac{eFL}{k_B T})^2]$  yields a linear dependence at small  $P$ , however, it does not fit the measured  $G(F)$  any better than the other models. It is unfortunate that neither model may be trusted to fit experiments on real systems except over a limited

range of fields. Given this situation, comparing the MW-enhanced conductance with the experimentally measured  $G(F)$  is the only option one has for a meaningful test.

The similarity between the field assisted and the MW-enhanced conductance and the qualitative agreement with theory make a strong case for the mechanism we propose. This is a plausible scenario for the MW frequency regime employed in this study as the associated wavelength is larger than the sample size.

#### IV. ACKNOWLEDGMENTS

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